Supporting Information

Carbon Nanotubes Produced from Ambient Carbon Dioxide for Environmentally Sustainable Lithium-Ion and Sodium-Ion Battery Anodes

Stuart Licht^{1,*} Anna Douglas², Jiawen Ren¹, Rachel Carter², Matthew Lefler¹, and Cary L. Pint^{2,*}

¹Department of Chemistry, George Washington University, Washington, DC 20052, USA

²Department of Mechanical Engineering, Interdisciplinary Materials Science Program, and Vanderbilt Institute of Nanoscale Science and Engineering, Vanderbilt University, Nashville TN 37235

*Corresponding Authors: slicht@gwu.edu, cary.l.pint@vanderbilt.edu

Experimental Details

Sodium Ion and Lithium Ion Battery Device Fabrication and Electrochemical Testing

CNTs produced using the STEP process, which is described in detail elsewhere, [1] were assembled into half-cell devices in an Ar glovebox using 2032 stainless steel coin cells (MTI). For the sodium ion batteries, the CNTs were combined in a slurry with conductive carbon black and PVDF binder at a ratio of 3:1:1, respectively. The slurry is assembled with a 2500 Celgard

separator saturated with 1 M NaPF₆ in diethylene glycol dimethyl ether (DGM) (Sigma-Aldrich) separating the cathode material from pure sodium foil (Sigma-Aldrich). The same procedure is followed for the lithium ion batteries, using lithium (Sigma-Aldrich) as the counter electrode and, for the electrolyte, 1 M LiPF₆ in 1 g/1 mL of ethylene carbonate (EC) and diethyl carbonate (DEC). Based on assessment of the electrodes pressed into coin cells, typical mass density loadings were observed to be ~ 0.06 mg/cm², and the density of the electrode with 60% CNT, 20% PVDF, and 20% carbon black was measured near ~ 0.54 g/cm³. The devices were tested utilizing a Metrohm Autolab multichannel testing system. Cyclic voltammetry was performed at room temperature on the devices between 0 and 3.0 V at a constant scan rate of 0.1 mV/s, and galvanostatic charge—discharge measurements were carried out for constant currents ranging from 0.1 to 1 A/g, but in most cases reported in the text involving rates of 0.1 A/g. Cycling studies were performed at 0.1 A/g.

Material Analysis: SEM and Raman Spectroscopy

To characterize materials used in batteries, both SEM and Raman spectroscopic analysis was performed. SEM was carried out using a Zeiss Merlin microscope using 5 kV accelerating voltage and working distance of ~ 4 mm. Raman measurements were performed on a Renishaw inVia microscope using 532 nm laser excitations.

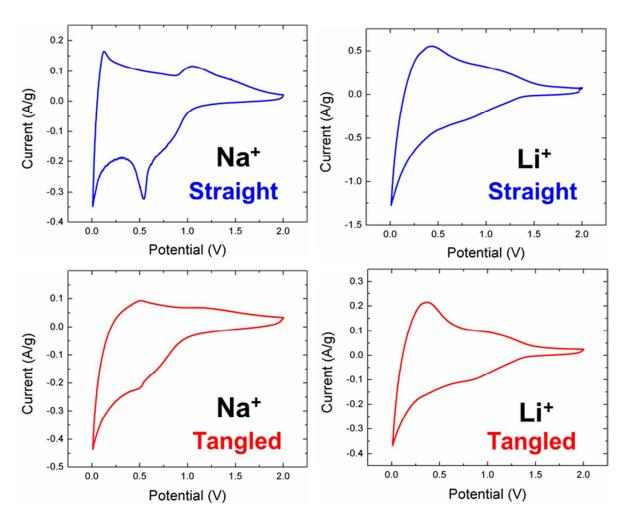


Figure S1. Cyclic voltammetry measurements at 100 mV/second for straight and tangled CNTs as anodes in both Li+ and Na+ cells. The low voltage insertion and extraction in all devices emphasizes anodic performance

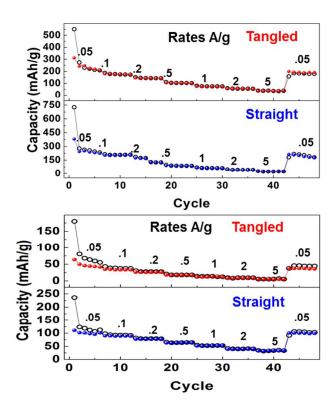


Figure S2. Rate capability studies for Li+ and Na+ cells incorporating CO₂-derived CNTs as anodes. Cycling rates (in A/g) are labeled on the plots.

References

[1] Ren, J. W.; Li, F. F.; Lau, J.; Gonzalez-Urbina, L.; Licht, S. One-Pot Synthesis of Carbon Nanofibers from CO2. *Nano Lett.* **2015**, *15*, 6142-6148.